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# Dynamics and thermodynamics of quantum crystals near the instability point in the self-consistent phonon theory

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## ABSTRACT

Formally one can distinguish between the thermodynamic stability conditions (the free energy of the lattice should have a minimum with respect to small variations mechanical or otherwise) and the dynamical ones (the phonon spectrum of the lattice should be positively defined). These last conditions are easily formulated in the self-consistent phonon theory (SCPT) based on the thermodynamic double-time Green's function method. According to it the dynamic instability temperature  $T_s$  for a simple Bravais lattice defines the temperature at which the bound crystalline state of atoms vanishes that really means that the phonon frequencies become complex at sufficiently high temperatures  $T \leq T_s$ .

Using the reduced second order approximation of the SCPT the dynamics of crystal lattice and the thermodynamical properties of the quantum crystals in the vicinity of the instability point are investigated. The results of calculations of the pressure dependence of the instability temperature, melting criterion, internal and free energy, free Gibbs energy as well as selected dynamic properties obtained with the help of the generalized form of the Buckingham, the Lennard-Jones and the Morse self-consistent potentials are given and compared with experimental data of solid h.c.p.  $^4\text{He}$  and f.c.c.  $^{20}\text{Ne}$ .

Comparison of the theoretical and experimental results allows us to state that the limiting temperatures of the dynamical stability obtained for the above-mentioned models pair potentials always appear to be the upper estimations of the real melting temperature.

**Keywords:** self-consistent phonon theory, quantum crystals, stability condition of solid helium and neon.

## 1. INTRODUCTION

In a series of papers<sup>1,2</sup> Plakida and Siklós have developed the self-consistent phonon theory (SCPT) of highly anharmonic crystals. This theory has been considered for some models of crystals<sup>3-6</sup>. In this paper, using the SCPT in the reduced second order approximation (RSOA) the dynamic and thermodynamic properties of quantum crystals in the vicinity of the instability point are investigated.

As it appears in the RSOA the limiting temperature of dynamical stability  $T_s^{dyn}$  may be obtained through a detailed analysis of the solvability of the self-consistent (s.c.) equation for the mean square relative displacement of neighbouring atoms:

$$\langle u^2(l) \rangle = \left\langle \left[ \tilde{u}_i - \tilde{u}_0 \right]^2 \right\rangle / l^2, \quad (1)$$

where  $\tilde{u}_0$  and  $\tilde{u}_i$  are the displacements of neighbouring atoms from their equilibrium positions and  $l = l(T, p)$  is their separation.  $\langle u^2(l) \rangle = \langle u^2(T, p) \rangle$  depends on the explicit form of the s.c. potential for the pairwise atomic interactions  $\tilde{\varphi}(l)$ . In particular, for the high temperature limit<sup>2</sup>:

$$\langle u^2(l) \rangle \approx \frac{6\theta}{zf(T, p)} \left\{ [1 - 0.11C_1(T, p)]^{-1} + C_2(T, p) \right\} \quad (2)$$

where:  $\theta = k_B T$ ,  $C_1(T, p) = \theta g^2(T, p) / f^3(T, p)$ ,  $C_2(T, p) = \hbar^2 f(T, p) / 3M\theta^2$ .

$f(T, p) = \partial^2 \tilde{\varphi} / \partial l^2$  and  $g(T, p) = \partial^3 \tilde{\varphi} / \partial l^3$  are the anharmonic force constants.  $z$ ,  $\hbar$ ,  $k_B$  and  $M$  are the number of nearest neighbours, Planck constant divided by  $2\pi$ , the Boltzmann constant and mass of atom, respectively.

The equation (2) has a different number of real solutions depending on the values of temperature  $T$  and pressure  $p$ . As it was shown by Siklós and Aksienov<sup>7</sup> for any given  $p$ , below a critical pressure  $p_c$ , and for  $T \leq T_s^{dyn}$  the lowest solution is the only physical one and corresponds to a stable crystal state.

In this paper we study the problem of lattice stability of quantum crystals (solid h.c.p.  $^4\text{He}$  and f.c.c.  $^{20}\text{Ne}$ ). Apart from the pressure variations of  $T_s^{dyn}$  we also investigate the behaviour of selected dynamic and thermodynamic properties of these crystals near the limit of stability. The detailed calculations are performed for the following s.c. potentials:

- exponential-power ( $exp, m$ ) Buckingham ( $B$ )

$$\tilde{\varphi}_B(l) = \frac{\alpha m}{\alpha - m} D_0 \left\{ \frac{1}{\alpha} \exp(\alpha x_1) - \sum_{k=0}^{\infty} \frac{1}{k!} y^k A_m^{(2k)} x^{m+2k} \right\}, \quad (3)$$

- purely power ( $n, m$ ) Lennard-Jones ( $LJ$ )

$$\tilde{\varphi}_{LJ}(l) = \frac{nm}{n-m} D_0 \sum_{k=0}^{\infty} \frac{1}{k!} y^k \left[ A_n^{(2k)} x^{n+2k} - A_m^{(2k)} x^{m+2k} \right], \quad (4)$$

- purely exponential ( $exp, exp$ ) Morse ( $M$ )

$$\tilde{\varphi}_M(l) = \frac{\alpha\beta}{\alpha - \beta} D_0 \left\{ \frac{1}{\alpha} \exp(\alpha x_1) - \frac{1}{\beta} \exp(\beta x_2) \right\}, \quad (5)$$

The symbols used in (3) to (5) have the following meaning:

$$x = \frac{r_0}{l}, \quad x_1 = \left( 1 - \frac{l}{r_0} + \alpha y \right), \quad x_2 = \left( 1 - \frac{l}{r_0} + \beta y \right), \quad y = \langle u^2(l) \rangle / 2r_0^2,$$

$$A_p^{(q)} = \begin{cases} \frac{1}{p'} & \text{for } q = 0 \\ \frac{1}{p'} p'(p'+1) \dots (p'+k-1) & \text{for } q = 1, 2, \dots \end{cases}$$

$n, m, \alpha$  and  $\beta$  are the dimensionless parameters describing the slope of the potentials curves,  $r_0$  and  $D_0$  define the potential minimum and are, respectively, the equilibrium distance of two isolated atoms and the energy of their dissociation. In order to obtain the numerical values of these parameters we take the zero point experimental data for the nearest-neighbours separation  $l_0$ , the energy of sublimation  $E_0$  and the isothermal compressibility  $K_0$  together with the relationships connecting these quantities with the interatomic interaction function.<sup>6</sup>

## 2. DYNAMIC AND THERMODYNAMIC FUNCTIONS OF STRONGLY ANHARMONIC QUANTUM CRYSTALS.

To discuss the dynamics and thermodynamics of the model we should calculate the pressure variation of the renormalization parameter  $y$ , the limiting temperature of dynamic stability  $T_s^{dyn}$ , the anharmonic force constants  $f(T, p)$  and  $g(T, p)$  as well as the internal energy  $E$ , the binding energy of lattice atoms  $D$ , the enthalpy  $H$ , the free energy  $F$  and the free Gibbs energy  $G$ . By taking into account only the renormalized cubic anharmonic interaction, we obtain the same expressions for above mentioned functions as in the conventional perturbation theory but with renormalized frequencies.<sup>2</sup> The general analytical forms for them are presented collectively in our previous paper<sup>8</sup> and are used in this paper in order to characterize the physical properties of h.c.p. helium and f.c.c. neon in numerical form with the help of the RSOA. The calculations are extended from low to high pressures.

### 3. NUMERICAL RESULTS.

Using the values of potential energy parameters given in Table 1 for the Buckingham (Eq.(3)), Lennard-Jones (Eq.(4)) and the Morse (Eq.(5)) s.c. functions we have found for the quantum crystals the pressure variations of the limiting temperature of dynamic stability  $T_s^{dyn}$  (Table 2). Numerical values of  $T_s^{dyn}$  are listed in Table 2 together with experimental data of the melting temperature  $T_m$  for comparison.

Table 1. Optimal values of the Buckingham (*B*), Lennard-Jones (*LJ*) and Morse (*M*) potential energy parameters of solid helium and neon calculated with the help of experimental data for the zero-point nearest-neighbour distance  $l_0$ , energy of sublimation  $E_0$  and isothermal compressibility  $K_0$ .<sup>9</sup>

Solids	Experimental data	Potential functions	Parameters					
			$D_0$ ( $10^{-23}$ J/atom)	$r_0$ (nm)	$\alpha$	$\beta$	$n$	$m$
<sup>4</sup> He h.c.p.	$l_0=0.36378$ nm	( <i>B</i> )	9.2250	0.2892	4.2	-	-	6.0
	$E_0=-0.07976 \cdot 10^{-21}$ J/atom	( <i>LJ</i> )	10.5325	0.2860	-	-	10.2	5.1
	$K_0=3.29 \cdot 10^{-8}$ Pa <sup>-1</sup>	( <i>M</i> )	9.3440	0.2692	6.0	3.0	-	-
<sup>20</sup> Ne f.c.c.	$l_0=0.31560$ nm	( <i>B</i> )	76.5	0.3001	19.0	-	-	6.0
	$E_0=3.115 \cdot 10^{-21}$ J/atom	( <i>LJ</i> )	77.5	0.2990	-	-	18.50	6.0
	$K_0=0.09 \cdot 10^{-8}$ Pa <sup>-1</sup>	( <i>M</i> )	73.8	0.3025	14.1	6.1	-	-

Table 2. Pressure variations of the limiting temperature of the dynamical  $T_s^{dyn}$  stability of solid helium and neon in the RSOA. Experimental values of the melting temperature  $T_m^{10,11}$  are given for comparison. The meaning of symbols is as in Table 1.

$p$	$T_m$	$T_s^{dyn}$ (K) for <sup>4</sup> He			$p$	$T_m$	$T_s^{dyn}$ (K) for <sup>20</sup> Ne		
(MPa)	(K)	(B)	(LJ)	(M)	(MPa)	(K)	(B)	(LJ)	(M)
	(Ex)					(Ex)			
3.222	1.86	10.9	8.2	8.7	0	24.6	25.9	26.8	26.0
4.164	2.13	11.8	8.8	9.9	10	26.0	27.5	28.6	27.5
5.248	2.41	12.6	10.7	11.0	20	27.5	29.1	30.4	29.0
6.414	2.69	13.5	12.2	12.0	32	29.2		32.8	
7.842	3.00	14.3	13.2	13.1	40	30.3	32.4	x	32.1
9.646	3.37	15.4	14.6	14.2	50	31.7	34.1	x	33.6
11.774	3.80	16.7	15.7	15.3	58	32.9	35.4	x	
12.256	4.27	18.3	17.7	16.6	63	33.6	x	x	x

Within the investigations of the lattice stability we have also computed the pressure variations of the modified melting criterion defined as the ratio  $x = \sqrt{\langle u^2 \rangle} / l_s$  ( $l_s$  – the equilibrium interatomic spacing at  $T = T_s^{dyn}$ ) (Table 3). It should be noted that this coefficient is, in the RSOA, the equivalent of the classical Lindemann criterion<sup>12</sup>.

The most characteristic results of our investigations of the dynamics and thermodynamics of the quantum crystals are given and compared with the available experimental data<sup>10,11,13</sup> in Tables 3-6.

Table 3. The effect of pressure on the distance between neighbouring atoms  $l$ , renormalization parameter  $y$ , melting criterion  $x$  and the anharmonic force constants  $f$  and  $g$  of solid helium. Experimental ( $l_{exp}$ ) values of  $l$  at  $T=0K^{10}$  are shown for comparison. The meaning of symbols is as in table 2.

<sup>4</sup> He h.c.p.		(B)					(LJ)					(M)				
$p$	$l_{exp}$	$l$	$y$	$x$	$f$	$g$	$l$	$y$	$x$	$f$	$g$	$l$	$y$	$x$	$f$	$g$
(MPa)	( $10^{-10}$ m)	( $10^{-10}$ m)	-	-	(N/m)	( $10^{10}$ Pa)	( $10^{-10}$ m)	-	-	(N/m)	( $10^{10}$ Pa)	( $10^{-10}$ m)	-	-	(N/m)	( $10^{10}$ Pa)
3.222	3.6378	3.6376	0.0314	0.1992	0.031	-0.139	3.6380	0.0336	0.2039	0.045	-0.271	3.6386	0.0439	0.2193	0.015	0.048
4.164	3.6080	3.6023	0.0298	0.1959	0.033	-0.147	3.6083	0.0316	0.1996	0.048	-0.285	3.5517	0.0410	0.2171	0.017	0.052
5.248	3.5777	3.5662	0.0282	0.1927	0.035	-0.156	3.5776	0.0298	0.1953	0.050	-0.301	3.4750	0.0385	0.2150	0.019	0.057
6.414	3.5468	3.5317	0.0269	0.1898	0.037	-0.165	3.5482	0.0281	0.1914	0.054	-0.318	3.4095	0.0365	0.2132	0.020	0.062
7.842	3.5155	3.4944	0.0254	0.1867	0.040	-0.177	3.5161	0.0264	0.1872	0.057	-0.338	3.3445	0.0345	0.2114	0.022	0.067
9.646	3.4835	3.4534	0.0240	0.1834	0.043	-0.190	3.4806	0.0247	0.1827	0.062	-0.364	3.2782	0.0325	0.2095	0.024	0.072
11.774	3.4509	3.4117	0.0226	0.1801	0.047	-0.206	3.4442	0.0230	0.1784	0.067	-0.393	3.2147	0.0307	0.2076	0.027	0.079
14.256	3.4178	3.3699	0.0212	0.1769	0.051	-0.223	3.4076	0.0215	0.1714	0.073	-0.426	3.1541	0.0291	0.2058	0.029	0.085

Table 4. Pressure variations of the internal energy  $E$ , binding energy  $D$ , enthalpy  $H$ , free Gibbs energy  $G$  and the free energy  $F$  (all quantities in units of  $10^{-21}$  J/atom) of <sup>4</sup>He obtained in the RSOA. Experimental data ( $E_{exp}$ ) for  $E$  at  $T=0K^{10}$  are given for comparison. The meaning of symbols is as in Table 2.

<sup>4</sup> He		(B)					(LJ)				
$p$	$E_{exp}$	$E$	$D$	$H$	$G$	$F$	$E$	$D$	$H$	$G$	$F$
(MPa)											
3.222	-0.07976	-0.07970	-0.04956	0.02996	-0.06539	-0.17505	-0.07964	-0.05927	0.03006	-0.28662	-0.39632
4.164	-0.07672	-0.07087	-0.04978	0.06678	-0.02093	-0.15858	-0.07772	-0.05937	0.06061	-0.23163	-0.36997
5.248	-0.07286	-0.06114	-0.05009	0.10718	0.02653	-0.14180	-0.07415	-0.05955	0.09578	-0.17369	-0.34364
6.414	-0.06803	-0.05111	-0.05046	0.14868	0.07414	-0.12565	-0.06922	-0.05981	0.13337	-0.11629	-0.31888
7.842	-0.06210	-0.03928	-0.05094	0.19734	0.12881	-0.10782	-0.06216	-0.06015	0.17890	-0.05118	-0.29224
9.646	-0.05492	-0.02498	-0.05155	0.25594	0.19338	-0.08754	-0.05228	-0.06062	0.23532	0.02477	-0.26283
11.774	-0.04581	-0.00881	-0.05225	0.32180	0.26472	-0.06589	-0.03984	-0.06118	0.30033	0.10772	-0.23245
14.256	-0.03533	0.00928	-0.05302	0.39505	0.34295	-0.04282	-0.02479	-0.06182	0.37410	0.19774	-0.20115

<sup>4</sup> He		(M)				
$p$	$E_{exp}$	$E$	$D$	$H$	$G$	$F$
(MPa)						
3.222	-0.07976	-0.07970	-0.04238	0.03006	0.00622	-0.10354
4.164	-0.07672	-0.07933	-0.04467	0.05261	0.03139	-0.10055
5.248	-0.07286	-0.07726	-0.04672	0.07849	0.05949	-0.09626
6.414	-0.06803	-0.07390	-0.04848	0.10586	0.08867	-0.09109
7.842	-0.06210	-0.06882	-0.05024	0.13864	0.12315	-0.08431
9.646	-0.05492	-0.06153	-0.05203	0.17876	0.16493	-0.07536
11.774	-0.04581	-0.05219	-0.05374	0.22440	0.21206	-0.06453
14.256	-0.03533	-0.04074	-0.05536	0.27559	0.26458	-0.05174

Table 5. The effect of pressure on the distance between neighbouring atoms  $l$ , renormalization parameter  $\gamma$ , melting criterion  $x$  and the anharmonic force constants  $f$  and  $g$  of solid neon. The meaning of symbols is as in Table 2.

$^{20}\text{Ne}$		(B)					(LJ)					(M)				
$p$	$l$	$\gamma$	$x$	$f$	$g$		$l$	$\gamma$	$x$	$f$	$g$	$l$	$\gamma$	$x$	$f$	$g$
(MPa)	( $10^{-10}\text{m}$ )	-	-	(N/m)	( $10^{10}\text{Pa}$ )		( $10^{-10}\text{m}$ )	-	-	(N/m)	( $10^{10}\text{Pa}$ )	( $10^{-10}\text{m}$ )	-	-	(N/m)	( $10^{10}\text{Pa}$ )
0	3.1562	0.0020	0.0607	0.707	-5.955		3.1563	0.0020	0.0600	0.774	-7.276	3.1562	0.0021	0.0628	0.577	-3.851
10	3.1471	0.0020	0.0601	0.741	-6.198		3.1472	0.0019	0.0593	0.812	-7.582	3.1470	0.0021	0.0622	0.603	-3.991
20	3.1388	0.0019	0.0595	0.774	-6.432		3.1388	0.0019	0.0587	0.848	-7.879	3.1385	0.0020	0.0617	0.628	-4.127
32	3.1296	0.0019	0.0588	0.812	-6.706		3.1296	0.0018	0.0580	0.891	-8.225	3.1290	0.0020	0.0611	0.657	-4.286
40	3.1238	0.0018	0.0584	0.837	-6.883		x	x	x	x	x	3.1230	0.0020	0.0607	0.677	-4.389
50	3.1171	0.0018	0.0579	0.868	-7.101		x	x	x	x	x	3.1159	0.0019	0.0602	0.700	-4.515
58	3.1119	0.0018	0.0575	0.892	-7.272		x	x	x	x	x	3.1105	0.0019	0.0599	0.719	-4.615
63	x	x	x	x	x		x	x	x	x	x	3.1073	0.0019	0.0597	0.730	-4.676

Table 6. Pressure variations of the internal energy  $E$ , binding energy  $D$ , enthalpy  $H$ , free Gibbs energy  $G$  and the free energy  $F$  (all quantities in units of  $10^{-21}$  J/atom) of  $^{20}\text{Ne}$  obtained in the RSOA. The meaning of symbols is as in Table 2.

$^{20}\text{Ne}$		(B)					(LJ)				
$p$		$E$	$D$	$H$	$G$	$F$	$E$	$D$	$H$	$G$	$F$
(MPa)											
0		-3.11548	-0.61877	-3.11548	-3.12680	-3.12680	-3.11417	-0.62026	-3.11417	-3.13319	-3.13319
10		-3.11860	-0.62232	-2.89820	-2.90859	-3.12899	-3.11847	-0.62414	-2.89805	-2.91575	-3.13617
20		-3.11968	-0.62552	-2.68236	-2.69193	-3.12925	-3.12062	-0.62765	-2.68329	-2.69983	-3.13716
32		-3.11870	-0.62900	-2.42513	-2.43385	-3.12742	-3.12085	-0.63145	-2.42725	-2.44258	-3.13618
40		-3.11688	-0.63114	-2.25468	-2.26288	-3.12508	x	x	x	x	x
50		-3.11347	-0.63362	-2.04270	-2.05032	-3.12108	x	x	x	x	x
58		-3.10991	-0.63547	-1.87396	-1.88114	-3.11710	x	x	x	x	x
63		x	x	x	x	x	x	x	x	x	x

$^{20}\text{Ne}$		(M)				
$p$		$E$	$D$	$H$	$G$	$F$
(MPa)						
0		-3.11568	-0.61358	-3.11568	-3.11855	-3.11855
10		-3.11684	-0.61637	-2.89645	-2.89889	-3.11927
20		-3.11609	-0.61891	-2.67888	-2.68094	-3.11815
32		-3.11305	-0.62168	-2.41986	-2.42151	-3.11470
40		-3.10989	-0.62338	-2.24835	-2.24977	-3.11130
50		-3.10482	-0.62538	-2.03522	-2.03636	-3.10596
58		-3.09996	-0.62687	-1.86567	-1.86660	-3.10089
63		-3.09658	-0.62777	-1.76011	-1.76063	-3.09740

The results of our investigations are collected in numerical forms, instead of the graphs, in order to complete the full experimental and theoretical characteristics of the properties of solid helium and neon.

#### 4. CONCLUSIONS

With the reduced, second order approximation<sup>2</sup> we have presented numerical calculations of the pressure variations of the limiting temperature of stability  $T_s^{dyn}$  (Table 2) and selected dynamic (Table 3 and 5) and thermodynamic properties (Table 4 and 6) of solid helium and neon. The calculations are performed in terms of the  $(exp,m)$  Buckingham, the  $(n,m)$  Lennard-Jones and the  $(exp,exp)$  Morse self-consistent potentials as interatomic potentials models in quantum solids.

The detailed analysis of the numerical values for  $T_s^{dyn}$ , (defined as the temperature at which the lattice as a bound state of the atoms due to the attractive part of the interatomic potential becomes unstable with respect to the collective excitations-self-consistency phonons<sup>2</sup>) show that ones are always the upper estimation of the real melting temperature – the better when the pressure is low and the pair potential hard. On the other hand the high-pressure results for the dynamical and thermodynamical quantities are generally better when the potential is soft. In connection with this we are convinced that, for further investigations of the quantum crystals, it is worthwhile taking all the three potentials but the RSOA of the lattice dynamics should be expanded by taking into account not only nearest-neighbour interactions but increasing over four the number of “important” terms in the potential energy decomposition. Work in this direction is in progress.

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